(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
1 August 2002 (01.08.2002)

PCT

(10) International Publication Number
WO 02/059212 A1

(51) International Patent Classification[7]: C08L 97/02, B27N 3/00 // (C08L 97/02, 89:00)

(21) International Application Number: PCT/EP02/00665

(22) International Filing Date: 21 January 2002 (21.01.2002)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
0101630.2 23 January 2001 (23.01.2001) GB

(71) Applicants *(for all designated States except US)*: AMYLUM EUROPE N.V. [BE/BE]; Burchtstraat 10, B-9300 Aalst (BE). ENSAM [FR/FR]; 2, Place P. Viala, F-34060 Montpellier (FR).

(72) Inventors; and
(75) Inventors/Applicants *(for US only)*: GUILBERT, Stéphane [FR/FR]; 3, Impasse de la Gerbe, F-34000 Montpellier (FR). REDL, Andréas [AT/BE]; Pont Zuid 47, B-9840 Zevergem/de Pinte (BE). MOREL, Marie-Hélène [FR/FR]; 44, Rue Pierre d'Auvergne, F-34000 Montpellier (FR).

(74) Agents: OSTYN, Frans et al.; K.O.B. N.V., Kennedypark 31c, B-8500 Kortrijk (BE).

(81) Designated States *(national)*: AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, OM, PH, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZM, ZW.

(84) Designated States *(regional)*: ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:
— *with international search report*
— *before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments*

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

WO 02/059212 A1

(54) Title: METHOD FOR PREPARING COMPOSITE MATERIALS CONTAINING NATURAL BINDERS

(57) Abstract: The invention relates to a method for preparing natural fibre-based composite materials containing natural binders and powdery proteins, comprising the following steps; adding a powdery protein adhesive binder to natural fibres having a moisture content of 1 15%; mixing the natural fibres together with the powdery protein adhesive; adjusting the moisture content of the composition to 6 24% w/w; and submitting the mixture to a heat pressure treatment to form the natural fibre-based material.

10/529459
JC06 Rec'd PCT/PTO 28 MAR 2005

METHOD FOR PREPARING COMPOSITE MATERIALS CONTAINING NATURAL BINDERS

The invention relates to a method for preparing natural fibre-based composite materials containing natural binders and powdery proteins.

With this methods, products are made in the form of artefacts such as panels and boards, composed of wood and/or non-wood natural fibre materials and a protein as the sole binder component (added).

Today, manufacture of the panels and boards is generally performed by means of hot pressing of wood or other vegetable fibres in presence of a reactive resin. Typical resins used during the mixing process are urea formaldehyde (UF) resins, phenol formaldehyde (PF) resins and melamine urea formaldehyde (MUF) resins. During the initial blending process aqueous resin is sprayed on the dry wood or vegetable fibres (2-4% moisture) and the whole is blended. The fibres dosed with resin can then be submitted to a further drying step. During this drying process the resin-dosed fibre is pneumatically or mechanically conveyed to the intermediate storage bins. From here, the fibre is transported to the mat forming line and to the pre-press. The thus formed mats are then entering the press and stacked. These mats are then submitted to hot pressing to consolidate the mat to a desirable panel density and thickness, to cure the resins and to heat stabilise the panel so that it will remain at the target thickness and density under normal service

conditions. Also continuous presses can be used. The thus obtained panels are then finished by shaving-off the edges by sanding the surfaces.
A disadvantage of this process is that the fibre material must be dried to a sufficient level to allow the blending with the liquid resin; otherwise homogeneous blending is not possible while lump formation occurs at too high moisture content.

An other disadvantage is that when adding the resin, the temperature during the blending and forming step must be controlled, because otherwise, a premature reaction occurs, and this is not desirable.

To reduce or avoid the use of the resin adhesives, the resin adhesives can be partially or fully replaced by renewable resources.

The use of animal or vegetable proteins in fibreboard is disclosed in Forest Products Journal (1998, vol.47, n°2, p.71-75), wherein the use of soya protein isolates in combination with synthetic resins is discussed. . Also the use of soya isolate powder is disclosed. In this method, wood particles are sprayed with 4% phenol formaldehyde resin, whereafter the mixture is further tumbled in a blender while 4% soya isolate powder is slowly added. Also the use of a soya isolate dispersion as the sole adhesive is disclosed.

However, the resultant fibreboard has the disadvantage that it has no moisture resistance.

The use of animal proteins in powder form is further disclosed in Sovjet patent application SU 1 813 640. In the disclosed method, lime milk (1,6-3,2%) and water (35-45%) are added to a fibre material at about 4% moisture content. In a next step, 8-16% albumin or casein powder glue is added, and the whole is mixed until the glue is uniformly distributed into the fibre material. The composition is then heated between 140-170°C at a pressure of 1,4-2,5 Mpa, this during 0,6-1,0 min/mm.

The use of wheat gluten as an adhesive has been disclosed in Starch (1968, vol20, n°12 p.395-399). The gluten that is used is reduced with sodium sulphite or thioglycolic acid.

The disadvantage related to these products is that the wheat gluten adhesive is obtained through a chemical reaction in an aqueous medium.

In Dutch patent application NL 1 003 133, the use of an adhesive based on wheat gluten for preparing fibreboard is disclosed. The wheat gluten glue used is a dispersion at 55-60% d.s., obtained by dispersing gluten into a solution of urea, citric acid and sodium bisulpite. The gluten glue, which is first mixed with a cross-linker (formaldehyde, glutardialdehyde or maleic anhydride), is then sprayed on and mechanically mixed with the wood fibre substrate.

The disadvantage here is that dispersing aids are needed to prepare the gluten glue dispersion.

Also the use of proteins as formaldehyde scavenger is disclosed in Dutch patent application NL 1 003 133, whereby 10% of a standard urea formaldehyde resin is replaced by gluten glue.

The disadvantage is that above 10% replacement of the standard urea formaldehyde resin, there is a clear loss of Internal Bond Strength.

In the European patent application EP 976 790, a process for the manufacture of composite materials is disclosed, in which a vegetable material containing fibre, or a mixture of such vegetable materials, are subjected to at least one thermoplastic processing step. The thermoplastic processing may be carried out in the presence of a bonding agent, for example a chemical bonding agent such as urea-formaldehyde, or a protein (which protein may be contained in the vegetable material or added to it). Prior to the thermoplastic processing step(s), the vegetable material(s) are subjected to at least one preliminary treatment. The product of the thermoplastic processing step(s) may be subjected to an after-treatment. The vegetable material(s) may be mixed with additives prior to and/or during the thermoplastic processing step(s). The composites may be formed in a variety of configurations, including board, sheets and films, and may find use as constructional items.

The disadvantage is that when proteins are used as the sole binder, relative to the total of the fibrous vegetable material and protein, the amount of water may be in the range of 25 to 50% by weight. As a result thereof, the compression-moulded articles must be submitted to an additional time and energy consuming drying step.

The purpose of the invention is to resolve the abovementioned disadvantages. This purpose is achieved by providing a method for preparing natural fibre-based composite materials containing natural binders and powdery proteins, comprising the following steps:

- adding a powdery protein adhesive binder to natural fibres having a moisture content of 1 - 15%;
- mixing the natural fibres together with the powdery protein adhesive;
- adjusting the moisture content of the composition to 6 - 24% w/w; and
- submitting the mixture to a heat pressure treatment to form the natural fibre-based material.

In a preferred method according the invention, the ratio of natural fibres and protein powder adhesive varies between 19:1 up to 1:1.

In a preferred method according the invention, the heat pressure treatment is performed within a temperature range of 100 - 250 °C.

In a preferred method according to the invention, the proteins added to the fibres have a moisture content varying between 4 and 14 % w/w.

In a more preferred method according the invention, the proteins added to the natural fibres have a moisture content between 8 and 12 % w/w.

In a specific method according to the invention, the wood or plant fibres have a moisture content between 1 and 20 % w/w.

In a more specific method according to the invention, the wood or plant fibres have a moisture content between 2 and 15 % w/w.

Preferably, the natural fibre-based composite materials include one of the products selected from packaging materials, decorative items, backing materials or structural materials.

In a preferred method according to the invention, said natural fibres can be obtained from whole plants or various parts thereof.

In a preferred method according to the invention, said natural fibres can be of animal origin.

In a preferred method according to the invention, said powdery protein adhesive can be of animal origin.

In another preferred method according to the invention, said powdery protein adhesive can be of vegetable origin.

In a preferred method according to the invention, the final moisture content of the composition is adjusted to 12 - 20% w/w.

In a more preferred method according to the invention, the final moisture content of the composition is adjusted to 14 - 18 % w/w.

In a preferred method according to the invention, the ratio of natural fibres and protein powder adhesive vary between 9:1 and 2:1.

In a more preferred method according the invention, the ratio of natural fibres and protein powder adhesive vary between 9:1 and 2,5:1.

In a preferred method according to the invention, the heat pressure treatment is performed within a temperature range of 175 - 225 °C.

In a specific method according to the invention, the heat pressure treatment is performed by means of compression moulding or by hot pressing in open presses.

In a specific method according to the invention, during hot pressing treatment a pressure is exercised that is sufficient to obtain a natural fibre-based composite

material with a density varying between 0,5 kg/dm³ and 1,5 kg/dm³.

In general, the term natural fibre-based composite materials concerns reconstituted products containing natural fibres originating from wood and/or annual plants, and an adhesive. Such composite materials include e.g. packaging materials, decorative items, backing materials, structural materials. More particularly, it refers to construction or building materials such as e.g. particleboard, medium density and high-density fibreboard, oriented strand board or chipboard. Other compositions comprise e.g. packaging materials (bottles, containers), decorative items (door panels), backing materials (carpet tiles, roofing materials), or structural materials (e.g. car bumpers).

Apart from wood, natural fibre materials can be obtained from whole plants or from various parts thereof. Textile fibres such as cotton, flax hemp or ramie can be used, but also meal products from the cereals or oilseed processing industry. Typical examples thereof are wheat bran, corn bran, wheat straw, barley husks, rapemeal, sunflower meal, soybean meal, etc. The natural fibre materials can also be of animal origin such as wool, silk or keratin-waste, etc.

The powdery protein adhesive can be of animal or vegetable origin. Animal protein sources are e.g. milk proteins, caseinates, whey concentrates and isolates, gelatine, fish proteins, egg albumin, plasma proteins,

animal flours, etc. The vegetable proteins can be selected among e.g. cereal proteins, tuberous proteins, proteins from leguminous origin, or oilseed proteins. Typical cereal proteins that can be used are wheat gluten, or maize gluten and derivatives thereof. Among the oilseed proteins, soy protein concentrates and isolates, rapeseed protein concentrates or sunflower protein concentrates, or derivatives thereof can be used, also. This list must not be considered as limiting, but merely as an illustration of the protein sources that can be used.

The proteins added to the natural fibres have a moisture content varying between 4 and 14% w/w, preferably varying between 8 and 12 % w/w. The wood or plant fibres may have a moisture content between 1 and 20 % w/w, 2 to 15% w/w being preferred. In contrast with standard conditions used during e.g. panel manufacture, the moisture content of the fibres can be higher because no additional moisture is added via the binding agent. This can offer an additional advantage with regard to drying cost reduction of the wood or plant fibres.

The natural fibres together with the powdery adhesive are mixed, and the final moisture content of the fibre/protein composition, before the heat pressure treatment, may vary between 6 and 24% w/w. Preferred ranges of moisture content are between 12 and 20% w/w, more preferably between 14 and 18% w/w. It is believed that his moisture content activates an initial sticking process, in which there occurs an interaction between the

proteins and the fibres. In this way, there is already a stabilisation before the heat pressure treatment. When the moisture content of the protein/fibre composition is too low, the phases can separate. When the moisture content of the fibre/protein composition is too high, more heat is needed to remove the water, and the final product has a less mechanical strength. With a moisture content between 6 and 24% w/w, the fibre/protein composition is already sufficiently homogeneous, which avoids an extra kneading step.

The ratio of natural fibre material and protein powder adhesive may vary between 19:1 up to 1:1, preferably between 9:1 and 2:1 and more preferably between 9:1 and 2,5:1.

Heat pressure treatment of the fibre/adhesive composition is performed within a temperature range of 100-250°C, preferably between 175°C and 225°C. Pressure exercised during hot pressing must be sufficient to obtain densities varying between 0,5 kg/dm³ and 1,5 kg/dm³.

The heat pressure treatment of the compositions may be performed by means of compression moulding or by hot pressing of the fibre/protein composition. The processing parameters, in particular temperature, pressure, and processing time will depend in any given case upon the nature of the starting materials and desired properties of the end products. It is observed that temperature evolution in the core of the product

during hot pressing or compression moulding is quite fast, as illustrated by figure 1 (see example 4).

The panel products obtained by means of the processes according to the invention do show excellent mechanical properties and sufficiently low water sensitivity.

It can be considered as an additional advantage that the prepared composites according to the invention are composed of biodegradable compounds. This can be of importance where materials are difficult to recycle.

The invention will now be further illustrated by way of specific examples, which are purely illustrative and not intended to limit the scope of the invention.

Example 1:

In this example the influence of the gluten fraction (GF), moisture content (MC) and pressing temperature (T), on the density (D) and the thickness swelling (TS) of panel boards, containing hemp fibres, is illustrated. Especially the thickness swelling (water sensitivity) is a measure for the quality of adhesion between fibres and matrix.

Materials used:

- vital wheat gluten, moisture content 8,6% (Amylum Aquitaine, Bordeaux, France)
- Hemp fibres, moisture content 8,9%, mean size 3 mm (Agricultural cooperative "La Chanvrière de l'Aube"

The hemp fibres are mixed with wheat gluten in a tumbling mixer (Heidolph Rheax 2, Germany) for 10 minutes. The volume of the mixing bowls is 100 ml, the total mass is 10g, the fibre weight fraction is 0.6, 0.7, 0.8, 0.9 w/w. Water is sprayed in the mixing bowl in order to raise the final water content to 18 %, the water content of the mixture without water addition being 9%.
The fibre/gluten mix is poured into a cylindrical mould (diameter 35 mm) and pressed for 10 minutes at 100, 125, 150, 175, and 200°C under 10T load.
The samples thus obtained are immersed in distilled water (at 25°C) for 24h. The thickness swelling is then determined as the percentage increase of the sample thickness as measured at the centre of the disks with a digital calliper square.
The material density is determined after measuring the sample thickness and diameter with a digital calliper square to the nearest 0,01mm and weighting the sample on a precision balance.
The moisture content of raw materials and samples is determined by weight difference after 24h drying at 104°C.

The results are given in table 1.

TABLE 1: Gluten/hemp fibre compositions: influence moisture content (MC) and pressing temperature (T) on density (D) and thickness swelling (TS)

Gluten fraction (w/w)	MC (%dm)	T (°C)	D (T/m³)	TS (%,24h)
0,3	8,65	125	1,43	114,25
0,3	8,65	150	1,44	41,21
0,3	8,65	175	1,32	31,25
0,3	16,9	125	1,41	28,21
0,3	16,9	150	1,33	18,73
0,3	16,9	175	1,23	18,38
0,3	16,9	200	1,12	19;55
0,4	17	175	1,06	40;06
0,2	17	175	1,17	54;08
0,1	17	175	1,28	78;09

Example 2:

In this example different types of natural fibres are used. The method of preparing them is analogous to the process described in example 1. Also here, density and thickness swelling are determined in function of pressing temperature and moisture content.

The following materials are used: wheat straw, wheat bran, and quebracho wood flour.

The gluten fraction is always 0,3. In table 2, the results of thickness swelling and density are given.

TABLE 2: Comparison of different fibres: influence moisture content (MC) and pressing temperature (T) on density (D) and thickness swelling (TS)

Fibre type	MC (%dm)	T (°C)	D (T/m³)	TS (%,24h)
Wheat straw	8,15	150	1,23	133,53
Wheat straw	8,15	175	0,98	35,89
Wheat straw	16,5	125	1,06	78,57
Wheat straw	16,5	175	1,00	28,38
Wheat straw	16,5	200	0,93	35,69
Quebracho flour	8,5	175	1,22	15,41
Quebracho flour	16,8	150	1,22	13,37
Quebracho flour	16,8	175	1,06	21,95
Wheat bran	9,9	150	1,09	27,38
Wheat bran	18,1	150	1,18	26,01

Example 3:

In this example, mechanical properties of gluten /fibre compositions are determined. The same method as in example 1 is used, except that the panels are formed in a rectangular mould of 120mm x 10mm. The thickness obtained is between 5 and 6 mm.

The mechanical properties are investigated according to norm EN ISO14125 in bending mode with TAXT2 texture analyser (Stable Microsystems, UK). The distance between flexion points is 100 mm. The samples are analysed directly after fabrication.

Materials used:

- vital wheat gluten, moisture content 8,6% (Amylum Aquitaine, Bordeaux, France)
- Hemp fibres, moisture content 8,9%, mean size 2 and 6 mm (Agricultural cooperative "La Chanvrière de l'Aube")
- Linen straw (8,9% MC, Unilin NV, B-8710 Wielsbeke)

The results of the mechanical testing are given in table 3.

TABLE 3: Influence of fibre type, moisture content (MC),pressing temperature (T) and gluten fraction (GF) on modulus of elasticity (MOE) and modulus of rupture (MOR). Mean values of 4 replicates.

Fibre type	MC (%dm)	T (°C)	GT (w/w)	MOE (GPa)	MOR (MPa)
Hemp 2 mm	9,3	150	0,2	3,63	31,51
Hemp 2 mm	18,0	150	0,2	2,54	33,34
Hemp 2 mm	9,3	175	0,2	3,48	26,14
Hemp 2 mm	18,0	175	0,2	3,22	31,27
Hemp 6 mm	18,0	150	0,3	3,22	33,91
Hemp 6 mm	18,0	150	0,2	3,62	32,99
Hemp 6 mm	18,0	150	0,1	2,44	29,82
Linen straw	9,0	175	0,2	4,53	43,17
Linen straw	18,0	150	0,2	2,75	25,56

Example 4:

Materials

Vital wheat gluten, (8.6% Moisture content, Amylum Aquitaine, Bordeaux, Fr)
Wood particles (8.5 % MC, Unilin NV, Schaapdreef 36, B-8710 Wielsbeke)

Methods

- Mixing: The fibres are mixed with wheat gluten in a 5 l rectangular mixing bowl by hand for 10 min, simulating the movements of a tumbling mixer.
- Pressing: A rectangular fibre mat of dimensions 17x26 cm and about 4cm height is formed by hand using a wooden frame. A k type thermocouple is placed in the centre of the fibre mat. After removal of the wooden frame, the mat is placed into the heated press (regulated at 175°C) and the temperature recording is started. The mat is compressed to 11 mm final thickness in a hand pump hydraulic press. The time needed for press closure to 11 mm is 60s, the load is maintained for 120s, then the load is relaxed gradually to zero during 1 min. The total pressing time is 4 min. 4 flexion specimens (30*170 mm) and 2 specimens (50*50 mm) for thickness swelling and determination of density are cut of each board.

The mechanical properties are investigated in bending mode with a ZWICK 500N universal testing machine. The distance between flexion points is 100 mm, cross-head

speed is 2mm/min. The samples are tested immediately after their fabrication.

Thickness swelling is investigated according to norm EN 319. The test specimens are immersed in distilled water (20°C), thickness is determined in the centre of the test specimen after 2 and 24h of immersion.

Results

- Core temperature evolution

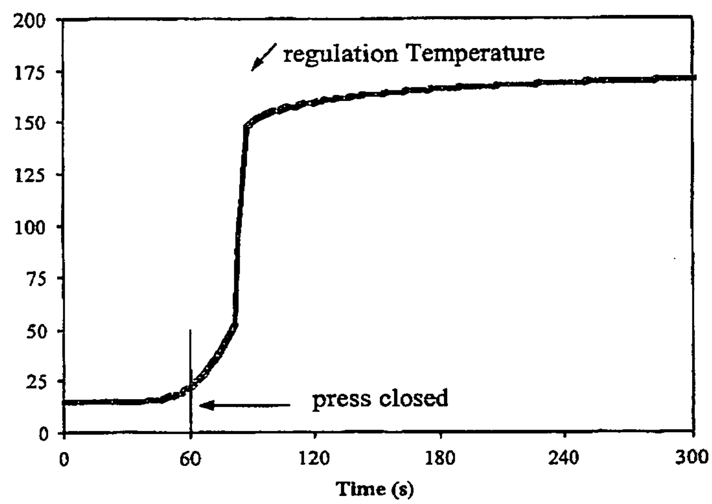

FIGURE 1: Core temperature evolution during pressing of gluten and wood particles. 175°`C regulation temperature, 300g wood (9%MC), 60g gluten (7%MC)

Figure 1 shows the core temperature evolution during the fabrication of a gluten wood particleboard. It is

observed that temperature rises very rapid as soon as the press is closed and the nominal thickness is reached. Core temperature reaches 150°C in 30 sec after press closure (90s total press time).

- Mechanical properties

TABLE 4: Mechanical properties and water sensitivity of gluten wood particleboards.

Sample N°		**E**	**F**	**G**	**H**	**Specifications**
Composition						EN 312-2
Fibres	g	300	300	300	300	
Gluten	g	60	75	75	33,3	
Thickness	mm	11,4	11,5	11,2	11,7	
Density	kg/m³		762,4	693,3	553	
Mechanical properties						
MOE	Gpa	0,976	1,029	1,077	0,636	
sdev		0,094	0,188	0,201	0,184	
MOR	Mpa	10,58	12,26	12,29	5,94	12,5
sdev		10,58	2,78	2,59	1,04	
Thickness swelling						
TS (2h)			48,8	52,2	40,5	
sdev			3,9	6	2,6	
TS (24h)			60,3	63,9	53,6	
sdev			1,7	1,9	0,1	

MOE is modulus of elasticity, MOR is modulus at break, TS is thickness swelling after immersion in 20°C water.

Specifications are: EN 312-2,general purose particleboard

The production of gluten wood particleboard is feasible. Until now boards with a glut.n weight fraction of 0.2 comply with the specifications of general purpose particleboard (EN 312-2).

Example 5:

In this example mechanical properties of different protein /fibre compositions are determined. The same method as in example 1 is used, except that the panels were formed in a rectangular mould of 120mm x 10mm. Moulding temperature is 175°C and moisture content is 9%. The fibre fraction is 0,8. Thickness obtained is between 5 and 6 mm.
Mechanical properties are investigated according to norm EN ISO 14125 in bending mode with TAXT2 texture analyser (Stable Microsystems, UK). The distance between flexion points is 100 mm. The samples are analysed directly after fabrication. The results are displayed in table 5.

Materials used:

- vital wheat gluten, moisture content 8,6% (Amylum Aquitaine, Bordeaux, France)
- Linen straw (8,9% MC, Unilin NV, B-8710 Wielsbeke)
- Zein (8% MC, Sigma Chemicals)
- Corn gluten meal (8,5% MC)
- Soy protein isolate (7,8% MC PTI)

TABLE 5: Modulus of elasticity (MOE) and modulus of rupture (MOR) of protein/linen straw composite materials, moulded at 175°C, 9%MC, and 10 min press time:

Protein binder	MOE (Gpa)	MOR (Mpa)
Corn gluten meal	4,9	26
Zein	4,8	33
Soy protein isolate	5,6	31
Wheat gluten	5,0	38

Example 6:

This example illustrates the effect of the pressing temperature on the mechanical properties of the fibre boards prepared with wheat gluten as the sole binder.

The boards in this example are prepared by mixing 300g wood fibres (2% moisture) with 33,3g wheat gluten (7% moisture) in a T-bar rotating mixer for 3 minutes. Then 25g water, mixed with 5,5g parrafin emulsion was added and mixed for an additional 7 minutes.

The pressing procedure as described in example 4 is slightly modified, whereby the pressing temperature was increased to values of 200°C and 225°C and pressing time was 10s/mm. Density of the boards thus prepared varied between 0,732 and 0,735.

The boards thus obtained were submitted to mechanical testing, as already described in example 4.

Pressing T	Modulus of elasticity (GPa)	Internal bond (MPa)	Thickness swelling in water (%)
175°C	1,12	0,31	94,8
200°C	1,36	0,40	100,3
225°C	1,50	0,61	71,4

CLAIMS

1. Method for preparing natural fibre-based composite materials containing natural binders and powdery proteins, comprising the following steps:
   - adding a powdery protein adhesive binder to natural fibres having a moisture content of 1 - 15%;
   - mixing the natural fibres together with the powdery protein adhesive;
   - adjusting the moisture content of the composition to 6 - 24% w/w; and
   - submitting the mixture to a heat pressure treatment to form the natural fibre-based material.

2. Method according to claim 1, **characterised in that** the ratio of natural fibres and protein powder adhesive varies between 19:1 up to 1:1.

3. Method according to any one of claims 1 and 2, **characterised in that** the heat pressure treatment is performed within a temperature range of 100 - 250 °C.

4. Method according to any one of claims 1 up to and including 3, **characterised in that** the proteins added to the natural fibres have a moisture content varying between 4 and 14 % w/w.

5. Method according to claim 4, **characterised in that** the proteins added to the fibres have a moisture content between 8 and 12 % w/w.

6. Method according to any one of claims 1 up to and including 5, **characterised in that** the wood or plant fibres have a moisture content between 1 and 20 % w/w.

7. Method according to any one of claims 1 up to and including 6, **characterised in that** the wood or plant fibres have a moisture content between 2 and 15 % w/w.

8. Method according to any one of claims 1 up to and including 7, **characterised in that** the natural fibre-based composite materials include one of the products selected from packaging materials, decorative items, backing materials or structural materials.

9. Method according to any one of claims 1 up to and including 8, **characterised in that** said natural fibres can be obtained from whole plants or various parts thereof.

10. Method according to any one of claims 1 up to and including 8, **characterised in that** said natural fibres can be of animal origin.

11. Method according to any one of claims 1 up to and including 10, **characterised in that** said powdery protein adhesive can be of animal origin.

12. Method according to any one of claims 1 up to and including 10, **characterised in that** said powdery protein adhesive can be of vegetable origin.

13. Method according to any one of claims 1 up to and including 11, **characterised in that** the final moisture content of the composition is adjusted to 12 - 20% w/w.

14. Method according to claim 13, **characterised in that** the final moisture content of the composition is adjusted to 14 - 18 % w/w.

15. Method according to any one of claims 1 up to and including 14, **characterised in that** the ratio of natural fibres and protein powder adhesive vary between 9:1 and 2:1.

16. Method according to claim 15, **characterised in that** the ratio of natural fibres and protein powder adhesive vary between 9:1 and 2,5:1.

17. Method according to any one of claims 1 up to and including 16, **characterised in that** the heat pressure treatment is performed within a temperature range of 175 - 225

18. Method according to any one of claims 1 up to and including 17, **characterised in that** the heat pressure treatment is performed by means of compression moulding or by hot pressing in open presses.

19. Method according to any one of claims 18, **characterised in that** during hot pressing a pressure is exercised that is sufficient to obtain a natural fibre-based composite material with a density varying between 0,5 kg/dm³ and 1,5 kg/dm³.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/EP 02/00665

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C08L97/02 B27N3/00 //(C08L97/02,89:00)

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, PAJ, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	PATENT ABSTRACTS OF JAPAN vol. 1998, no. 10, 31 August 1998 (1998-08-31) & JP 10 113913 A (MATSUYAMA KANKYO SERVICE:KK), 6 May 1998 (1998-05-06) abstract	1-9, 12-19
X	US 2 543 925 A (A.M.MONATH) 6 March 1951 (1951-03-06) column 1, line 1 - line 5; claims claims column 5, line 7 - line 17 column 3, line 42 - line 70 column 3, line 20 - line 29 column 2, line 16 - line 17	1-11, 13-19

-/--

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
26 April 2002	11/06/2002

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Mazet, J-F

INTERNATIONAL SEARCH REPORT

Int onal Application No
PCT/EP 02/00665

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 556 577 A (KRICKL LUDWIG) 25 August 1993 (1993-08-25) column 1, line 41 - line 48 claims 1-5,8,10,12-18,20,22 column 4, line 18 - line 23	1-19
A	US 5 360 586 A (WYATT ET AL.) 1 November 1994 (1994-11-01) abstract column 2, line 37 - line 48 column 3, line 9 - line 18 example	1
A	US 1 427 378 A (P. GRUBER ET AL.) 29 August 1922 (1922-08-29) claims	1
A	WO 98 06785 A (SILBIGER J.) 19 February 1998 (1998-02-19) claims	1
A	EP 0 987 089 A (TOULOUSAINE DE RECHERCHE ET DE DEVELOPPEMENT T.R.D. ANONYME) 22 March 2000 (2000-03-22) page 3, line 11 - line 25 page 5, line 55 -page 6, line 27; example 3 claims 1-3,6,12,13 page 3, line 49 - line 53	1-9, 12-14, 17-19
A	WO 96 14970 A (VALTION TEKNILLINEN TUTKIMUSKESKUS) 23 May 1996 (1996-05-23) page 2, line 8 - line 12 page 5, line 21 - line 33 example 1 page 5, line 9 - line 13	1,2,4-9, 12-16
A	DE 198 43 493 A (IHD INSTITUT FÜR HOLZTECHNOGIE DRESDEN GMBH) 23 March 2000 (2000-03-23) page 4, line 30 - line 48; claims	1-9, 12-16, 18,19
A	DATABASE WPI Week 199527 Derwent Publications Ltd., London, GB; AN 1995-201039 XP002197694 "Making method of rice straw plate" & CN 1 085 487 A (SHUANGQIAO RICE STRAW PLATE FACTIRY BEIJ), 20 April 1994 (1994-04-20) abstract	

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT/EP 02/00665

Patent document cited in search report		Publication date	Patent family member(s)		Publication date
JP 10113913	A	06-05-1998	NONE		
US 2543925	A	06-03-1951	NONE		
EP 556577	A	25-08-1993	DE	9200610 U1	19-11-1992
			DE	9208683 U1	01-10-1992
			EP	0556577 A1	25-08-1993
US 5360586	A	01-11-1994	NONE		
US 1427378	A	29-08-1922	NONE		
WO 9806785	A	19-02-1998	AT	209669 T	15-12-2001
			WO	9806785 A1	19-02-1998
			DE	59705574 D1	10-01-2002
			DK	918825 T3	25-03-2002
			EP	0918825 A1	02-06-1999
			US	6284838 B1	04-09-2001
EP 987089	A	22-03-2000	FR	2784047 A1	07-04-2000
			EP	0987089 A1	22-03-2000
WO 9614970	A	23-05-1996	FI	945337 A	12-05-1996
			AU	3873295 A	06-06-1996
			WO	9614970 A1	23-05-1996
DE 19843493	A	23-03-2000	DE	19843493 A1	23-03-2000
CN 1085487	A	20-04-1994	NONE		